

| | | | |
|--|---------------------------------------|---|---|
| FORM PTO-1390 (REV 5-93) | | U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE | ATTORNEY'S DOCKET NUMBER |
| TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371 | | | BAUER 214-KFM |
| | | | U.S. APPLICATION NO. (If known, see 37 CFR 1.5) |
| | | | 09/622301 |
| INTERNATIONAL APPLICATION NO. PCT/EP99/01036 | INTERNATIONAL FILING DATE 17/02/99 | PRIORITY DATE CLAIMED 18/02/98 | |
| TITLE OF INVENTION TUBULAR REACTOR FOR CATALYTIC REACTIONS | | | |
| APPLICANT(S) FOR DO/EO/US GUETLHUBER, FRIEDRICH | | | |
| Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information: | | | |
| <ol style="list-style-type: none"> <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371. <input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1). <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2)) <ol style="list-style-type: none"> <input checked="" type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau). <input type="checkbox"/> has been transmitted by the International Bureau <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US) <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. 371(c)(2)). <input checked="" type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3)) <ol style="list-style-type: none"> <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau). <input type="checkbox"/> have been transmitted by the International Bureau. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired. <input checked="" type="checkbox"/> have not been made and will not be made. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)). <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)). <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). | | | |
| Items 11. to 16. below concern other document(s) or information included: | | | |
| 11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98 | | | |
| 12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included. | | | |
| 13. <input checked="" type="checkbox"/> A FIRST preliminary amendment. <input type="checkbox"/> A SECOND or SUBSEQUENT preliminary amendment. | | | |
| 14. <input type="checkbox"/> A substitute specification. | | | |
| 15. <input type="checkbox"/> A change of power of attorney and/or address letter. | | | |
| 16. <input type="checkbox"/> Other items or information: | | | |

534 Rec'd PCT/PTO 16 AUG 2000

| | | | | | |
|--|--|------------------------------|--|--------------------------|--|
| U.S. APPLICATION NO (if known, see 37 CFR 1.5) | | INTERNATIONAL APPLICATION NO | | ATTORNEY'S DOCKET NUMBER | |
| 09/622301 | | PCT/EP99/01036 | | BAUER 214 | |

| | | | | | | | |
|--|--------------|--------------|---------|---------------|--|--------------|--|
| 17. <input checked="" type="checkbox"/> The following fees are submitted: | | | | CALCULATIONS | | PTO USE ONLY | |
| Basic National Fee (37 CFR 1.492(a)(1)-(5)): Search Report has been prepared by the EPO or JPO \$840.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) \$ 670.00 No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2)).. \$ 760.00 Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO..... \$ 970.00 International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4) \$ 96.00 ENTER APPROPRIATE BASIC FEE AMOUNT = | | | | 840.00 | | | |
| Surcharge of \$130.00 for furnishing the oath or declaration later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(e)) | | | | \$ | | | |
| Claims | Number Filed | Number Extra | Rate | | | | |
| Total Claims | 11 -20 = | 0 | X 18.00 | \$ | | | |
| Independent Claims | 1 -3 = | | X 78.00 | \$ | | | |
| Multiple dependent claims(s) (if applicable) | | | | +260.00 | | | |
| TOTAL OF ABOVE CALCULATIONS = | | | | \$ 840.00 | | | |
| Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity statement must also be filed. (Note 37 CFR 1.9, 1.27, 1.28). | | | | \$ 420.00 | | - | |
| SUBTOTAL = | | | | \$ 420.00 | | | |
| Processing fee of \$130.00 for furnishing the English translation later than <input type="checkbox"/> 20 <input type="checkbox"/> 30 months from the earliest claimed priority date (37 CFR 1.492(f)). | | | | + | | | |
| TOTAL NATIONAL FEE = | | | | \$ 420.00 | | | |
| Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40.00 per property + | | | | \$ | | | |
| TOTAL FEES ENCLOSED = | | | | \$ 420.00 | | | |
| | | | | Amount to be: | | | |
| | | | | refunded \$ | | | |
| | | | | charged \$ | | | |

a. ☒ A check in the amount of \$ 420.00 to cover the above fees is enclosed.

b. ☐ Please charge my Deposit Account No. _____ in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. ☒ The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 50-0427. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

Express Mail mailing label
number EK 677 256 499 US

Date of Deposit August 16, 2000

I hereby certify that this paper or fee is being deposited with the United States Postal Service Express Mail Post Office to _____ Service under 37 CFR 1.10 on the date indicated above and is addressed to the Commissioner of Patents and Trademarks, Washington, DC 20251.

SEND ALL CORRESPONDENCE TO:
MILDE, HOFFBERG & MACKLIN, LLP
10 BANK STREET
SUITE 460
WHITE PLAINS, NEW YORK 10606
914-949-3100

By: [Signature]
NAME Karl F. Milde, Jr.
REGISTRATION NUMBER 24,822

09/622301

534 Rec'd PCT/PTO 16 AUG 2000

BAUER 214-KFM:jg
D-10826

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : JOHANN ORTNER and JOHANN SEEMANN
Serial No.: TO BE ASSIGNED
Filed : HERewith
For : TUBULAR REACTOR FOR CATALYTIC REACTIONS

August 16, 2000

Hon. Commissioner of Patents
& Trademarks
Washington, DC 20231

Sir:

PRELIMINARY AMENDMENT

Prior to examination, please amend the above-identified
patent application as follows:

IN THE TITLE:

Please change the title of this application to
-- TUBULAR REACTOR FOR CATALYTIC GAS PHASE REACTIONS --.

IN THE SPECIFICATION:

On page 1, after the title, insert the following
heading:

-- BACKGROUND OF THE INVENTION --;

line 2, after "reactions", delete "according to the concept
of patent claim 1".

On page 3, line 17, delete "which is" and line 18,
delete "the basis for the concept of claim 1,".

On page 4, after line 12, insert the following heading:

-- SUMMARY OF THE INVENTION --;

and delete lines 20-22 and insert the following paragraph:

-- This objective, as well as other objectives which will become apparent from the discussion that follows, are achieved, in accordance with the present invention, by an improvement wherein the two tube plates are anchored in an essentially known manner at their edges to the reactor jacket in a sealed manner and wherein the heat insulation zone includes a chamber containing a solid, liquid or gaseous heat insulation material or components that are stream-calming with respect to the heat carrier. --

On page 5, delete lines 11-13 and insert the following paragraph and heading:

-- For a full understanding of the present invention, reference should now be made to the following detailed description of the preferred embodiments of the invention as illustrated in the accompanying drawings.

BRIEF DESCRIPTION OF THE DRAWINGS --;

and lines 16, 19 and 22, change "," to -- . --.

On page 6, line 3, change ", and" to -- . -- and delete lines 7 and 8 and insert the following heading and paragraph:

-- DESCRIPTION OF THE PREFERRED EMBODIMENTS

The present invention will now be described with reference to Figs. 1-5 of the drawings. Identical elements

in the various figures are identified by the same reference numerals --.

On page 11, after the last line, insert the following paragraph:

-- There has thus been shown and described a novel tubular reactor for catalytic gas phase reactions which fulfills all the objects and advantages sought therefor. Many changes, modifications, variations and other uses and applications of the subject invention will, however, become apparent to those skilled in the art after considering this specification and the accompanying drawings which disclose the preferred embodiments thereof. All such changes, modifications, variations and other uses and applications which do not depart from the spirit and scope of the invention are deemed to be covered by the invention, which is to be limited only by the claims which follow. --

IN THE CLAIMS:

Before line 1, change "Patent Claims" to:

-- C L A I M S

What is claimed is: --

Please amend claims 1-9 as follows:

1. (Once Amended) A tubular reactor [(2)] for catalytic gas phase reactions with a heat carrier that circulates around a contact tube bundle [(8)] inside a reactor jacket [(10)],

the tube bundle extending between a tube plate [(4)] at the gas inlet side and a tube plate [(6)] at the reaction gas outlet side beginning or ending there and sealed to it, with gas inlet and gas outlet hoods [(12, 14)] spanning the face sides of the two tube plates, and with a heat insulation zone [(46)] that borders on the heat carrier side on the tube plate at the gas inlet side, *[characterized in that]* the improvement wherein the two tube plates [(4, 6)] are anchored in an essentially known manner at their edges to the reactor jacket [(10)] in a sealed manner, and [that] wherein the heat insulation zone [(46)] includes a chamber [(64)] containing a solid, liquid or gaseous heat insulation material or components [(84)] that are stream-calming with respect to the heat carrier.

2. (Once Amended) Tubular reactor [(2)] as set forth in claim 1, *[characterized in that]* wherein the heat insulation zone [(46)] exhibits a locally varying thickness.

3. (Once Amended) Tubular reactor [(2)] as set forth in claim 1 [or 2, *characterized in that*] , wherein the heat insulation zone (46) exhibits a locally varying structure.

4. (Once Amended) Tubular reactor [(2)] as set forth in [one of the previous claims, *characterized in that*] claim 1, wherein the heat insulation zone [(46)] is limited to

partial [zones, such as] tube-free areas [or the edge area] of the tube plate on the gas inlet side [(4), for example].

5. (Once Amended) Tubular reactor [(2)] as set forth in [one of the previous claims with] claim 1, having a heat insulation zone [(46)] in the form of a chamber (64), [*characterized in that*] and wherein a liquid or gaseous heat insulation material in the chamber is prevented from circulating by structures that are installed in the chamber.

6. (Once Amended) Tubular reactor [(2)] as set forth in [one of the claims 1 through 4 with] claim 1, having a heat insulation zone [(46)] in the form of a chamber [(64)], [*characterized in that*] the improvement wherein a liquid or gaseous heat insulation material is circulated throughout the chamber as cooling medium.

7. (Once Amended) Tubular reactor [(2)] as set forth in claim 6, [*characterized in that*] wherein a partial stream of the heat carrier circulating around the contact tube bundle [(8) finds use] is used as a liquid or gaseous heat insulation material.

8. (Once Amended) Tubular reactor [(2)] as set forth in [one of the claims 1 through 4 with] claim 1, having a heat insulation zone [(46)] formed by built-in components [(84)],

[characterized in that] wherein the components exhibit honeycomb or concentric ring structures.

9. (Once Amended) Tubular reactor [(2)] as set forth in claim 8, [characterized in that] wherein the components [(84)] are covered, at least on the side opposite the tube plate [(4)] on the gas inlet side[, and preferably sealed].

Please add the following new claims:

-- 10. Tubular reactor as set forth in claim 9, wherein the components are sealed on the side opposite the tube plate on the gas inlet side.

11. Tubular reactor as set forth in claim 1, wherein the heat insulation zone is limited to the edge area of the tube plate on the gas inlet side. --

IN THE ABSTRACT:

Please add the ABSTRACT OF THE DISCLOSURE attached herewith on a separate sheet.

R E M A R K S

This Preliminary Amendment is being filed to place the specification and claims in proper form under United States

Patent Practice and also to add an Abstract. No new matter has been introduced.

Respectfully submitted,

By


Karl F. Milde, Jr.
Reg. No. 24,822

MILDE, HOFFBERG & MACKLIN, LLP
10 Bank Street - Suite 460
White Plains, NY 10606

(914) 949-3100

-- ABSTRACT OF THE DISCLOSURE

The invention relates to a tubular reactor (2) for catalytic reactions, which inside its reactor mantle (10) has a contact tube bundle (8) around which a heat carrying medium circulates. The tube bundle extends between a tube base on the reaction gas entry side and a tube base on the reaction gas exit side (4, 6; 60; 82). The invention also comprises gas entry and gas exit domes (12, 14) which span the two tube bases across their faces, and reaction-inhibiting means in the area of the tube base on the gas entry side. The invention is characterized in that the reaction-inhibiting means wholly or partly consist of a heat insulating layer (46; 50; 64; 80) which does not cover the tube in its cross-section and is situated on at least one side of the corresponding tube base (4; 60; 82). In this way either the corresponding tube base (4; 60; 82) is insulated against the hot heat carrying medium or the reaction gas entering the reactor is prevented from coming into contact with the relatively hot tube base, so that harmful secondary reactions at the reactor entry are avoided. --

Applicant or Patentee: FRIEDRICH GUETLHUBER Attorney's
Serial or Patent No.: TO BE ASSIGNED Docket No.: BAUER 214KFM
Filed or Issued: HEREWITH
For: TUBULAR REACTOR FOR CATALYTIC REACTIONS

VERIFIED STATEMENT (DECLARATION) CLAIMING SMALL ENTITY
STATUS (37 CFR 1.9(f) and 1.27 (b)) - INDEPENDENT INVENTOR

As a below named inventor, I hereby declare that I qualify as an independent inventor as defined in 37 CFR 1.9(c) for purposes of paying reduced fees under section 41(a) and (b) of Title 35, United States Code, to the Patent and Trademark Office with regard to the invention entitled TUBULAR REACTOR FOR CATALYTIC REACTIONS described in

☒ the specification filed herewith
☐ application serial no. _____ filed _____
☐ patent no. _____ issued _____

I have not assigned, granted, conveyed or licensed and am under no obligation under contract or law to assign, grant, convey or license, any rights in the invention to any person who could not be classified as an independent inventor under 37 CFR 1.9(c) if that person had made the invention, or to any concern which would not qualify as a small business concern under 37 CFR 1.9(d) or a nonprofit organization under 37 CFR 1.9(e).

Each person, concern or organization to which I have assigned, granted, conveyed, or licensed or am under an obligation under contract or law to assign, grant, convey, or license any rights in the invention is listed below:

☒ no such person, concern, or organization
☐ persons, concerns organizations listed below*

*NOTE: Separate verified statements are required from each named person, concern or organization having rights to the invention averring to their status as small entities. (37 CFR 1.27)

FULL NAME _____
ADDRESS _____
☐ INDIVIDUAL ☐ SMALL BUSINESS CONCERN ☐ NONPROFIT ORGANIZATION

FULL NAME _____
ADDRESS _____
☐ INDIVIDUAL ☐ SMALL BUSINESS CONCERN ☐ NONPROFIT ORGANIZATION

FULL NAME _____
ADDRESS _____
☐ INDIVIDUAL ☐ SMALL BUSINESS CONCERN ☐ NONPROFIT ORGANIZATION

I acknowledge the duty to file, in this application or patent, notification of any change in status resulting in loss of entitlement to small entity status prior to paying, or at the time of paying, the earliest of the issue fee or any maintenance fee due after the date on which status as a small entity is no longer appropriate. (37 CFR 1.28(b))

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application, any patent issuing thereon, or any patent to which this verified statement is directed.

| | | |
|-----------------------------|-----------------------|-----------------------|
| FRIEDRICH GUETLHUBER | | |
| NAME OF INVENTOR | NAME OF INVENTOR | NAME OF INVENTOR |
| <i>Friedrich Guetlhuber</i> | | |
| Signature of Inventor | Signature of Inventor | Signature of Inventor |
| <i>10. July 2000</i> | | |
| Date | Date | Date |

PTO/PCT Rec'd 25 OCT 2000

09/622301

TUBULAR REACTOR FOR CATALYTIC REACTIONS

The invention relates to a tubular reactor for catalytic reactions according to the concept of patent claim 1.

Usually, such reactors exhibit a reactor jacket containing a heat carrier that circulates around a contact tube bundle, which extends between a tube plate at the reaction gas inlet side and a tube plate at the reaction gas outlet side, as well as gas inlet and gas outlet hoods spanning the face sides of the two tube plates. The process gas, usually a gas mixture, that is to be brought to reaction enters a contact tube that contains a catalytic mass via the gas inlet hood and after passing said contact tube exits the reactor via the gas outlet hood. The gas inlet may be located either on the top or bottom side, and as a whole the heat carrier may pass through the reactor in parallel flow or counter flow with regard to the process gas flow. The reactor can also have a multi-step design as shown, for example, in DE 22 01 528 C, Fig. 5.

Usually, the process gas stream is comprised of two or more material streams that are combined shortly before entering the reactor, that is, its gas inlet hood. In this course, secondary reactions that are harmful to the process or even ignition and deflagration may occur, especially in the immediate surrounding of the generally relatively hot tube plate. Examples of such reaction processes are the production

of maleic acid anhydride, phthalic acid anhydride, acrolein and acrylic acid.

Fillers of ceramic materials or a wire mat mesh have been introduced into the gas inlet hood in an effort to prevent such secondary reactions. Other attempts consisted of insulating the tube endings by using cylinders, because the highest temperatures occur generally at the tube plate on the gas inlet side in the area of the tube ending. However, in the end, none of these measures proved effective or at least dependable for preventing the above mentioned secondary reactions.

For this reason, it is the objective of the invention to design a tubular reactor according to the concept such that secondary reactions, especially ignitions and deflagrations, are dependably prevented inside the gas inlet hood.

This objective is essentially achieved through the characteristic features of claim 1. The subclaims offer additional advantageous design options.

The respective heat insulation layer at the tube plate on the gas inlet side accomplishes on the gas inlet side that the entering process gas is kept from the hot tube plate and on the side of the heat carrier that circulates around the contact tubes and that the tube plate is kept cool in relation

to this heat carrier.

Preferred exemplary embodiments of the respective tubular reactor are described in greater detail below based on the drawings, wherein

Fig. 1 shows a schematic longitudinal section through a tubular reactor subject to the invention in a first embodiment together with connecting elements,

Fig. 2 shows a schematic longitudinal section through an end section at the gas inlet side of such a tubular reactor, however, in a variation,

Fig. 3 shows a schematic longitudinal section through an end section at the gas inlet side of a tubular reactor as in Fig. 1, however, in a different variation,

Fig. 4 shows a schematic longitudinal section through an end section at the gas inlet side of a tubular reactor subject to the invention in a different embodiment,

Fig. 5 shows a schematic longitudinal section through an end section at the gas inlet side of a tubular reactor subject to the invention in another embodiment, and

Fig. 6 shows a schematic longitudinal section through an

end section at the gas inlet side of a tubular reactor subject to the invention in yet another embodiment.

Easily comparable elements that occur in the individual Figures were given the same reference number.

The tubular reactor 2 shown in Fig. 1 features in typical manner a vertical tube bundle 8 that stretches inside a cylindrical reactor jacket 10 from a tube plate 4 at the gas inlet side to a tube plate 6 at the gas outlet side with the two tube plates 4 and 6 being attached to said cylindrical jacket in a sealed manner. The tube plate 4 at the gas inlet side is spanned by a gas inlet hood 12 and the tube plate 6 at the gas outlet side is spanned by a gas outlet hood 14, with both hoods being attached to the respective tube plate in a sealed manner. The individual tubes, e.g., 16, of the tube bundle 8 contain a gas-permeable catalytic mass. The reaction or process gas that is to be brought to a reaction is fed into the gas inlet hood 12 via a gas supply line 18, while the process gas that has passed through the tube bundle 8 and that has gone through the reaction process is removed from the gas outlet hood 14 via a gas removal line 20. Ring channels 22 and 24 surrounding the reactor jacket 10 close to the two tube plates 4 and 6 make possible the supply and removal of a heat carrier, which is in a liquid state at least when the reactor is in operation, into or from the reactor jacket 10, where said heat carrier circulates around the individual tubes,

e.g., 16, of the tube bundle 8 to remove the reaction heat. The flow of the heat carrier can be directed in a desired manner or, if required, distributed across the cross section of the reactor jacket as shown by using two types of guide plates 26 and 28. The heat carrier that is removed from the reactor jacket 10 via the ring channel 22 is returned to the reactor jacket 10 via the ring channel 24 by using a pump 30 whereby a controllable partial flow is guided through a cooling device (not shown here) via branch lines 32 and 34.

The process gas supplied to the reactor via the gas supply line 18 is (in this case) comprised of two partial streams 36 and 38 that are preheated using the heat exchangers 40 and 42, mixed in a mixing device 44 in order to be supplied to the gas inlet hood 12 via the gas supply line 18 as the aforementioned process gas.

In some cases, the supplied process gas is by itself already very reactive, especially if it has had contact with hot surfaces with the tube plate generally being the hottest surface within the space under the gas inlet hood 12. For this reason, the invention provides a heat insulation for the tube plate 4 either against the heat carrier that circulates around the tube bundle 8 as shown in Fig. 1, for example, or against the entering process gas as shown in Fig. 3, or both. In the one case, the insulation's effect is that the tube plate is kept comparatively cool and in the other case that the process

gas is prevented from contact with the hot tube plate.

According to Figures 1, 2 and 3, the heat insulation layer 46 that has been applied to the heat carrier side of the tube plate 4 consists of, for example, ceramics, such as a glass frit, or of a heat-resistant solid material and tightly surrounds the individual tubes, e.g., 16, just as tight as it connects to the reactor jacket 10. While the heat insulation layer 46 according to Fig. 1 has a consistent thickness, its thickness increases towards the center according to Fig. 2 under the assumption that the tube plate usually has the highest temperature in that area.

It is understood that for other temperature distributions, the profile of the heat insulation layer, e.g., 46, may have a different shape. For example, the heat insulation layer 46 can, as shown in Fig. 3, feature a collar 48 at the edge of the tube plate 4 along the interior wall of the reactor jacket 10 in order to keep the temperature gradient at the connection of the reactor jacket to the cooler tube plate, and thus, the temperature stress low. As can be seen from Fig. 3 as well, the thickness of the heat insulation layer 46 can also be different - generally greater - in the tube-free zones of the reactor than in the tube zones in order to accommodate for the normally greater heat of the tube plate in these zones. It is furthermore conceivable to vary the composition of the heat insulation layer instead of the thickness or the thickness of

the heat insulation layer alone, either by varying the amount ratio of its components or by selecting completely different materials. Furthermore, the heat insulation layer, such as 46, for example, can be limited to partial areas of the tube plate, for example, the tube-free zone or the edge area of the tube plate at the transition to the reactor jacket 10.

The same applies to the heat insulation layer 50 that is applied to the tube plate at the gas inlet side according to Fig. 4. It differs from the heat insulation layer 46 according to Figures 1 and 2, however, in that it must contain openings, such as, for example, 52, that correspond to and are in line with the interior cross-section of the individual tubes, such as, for example, 16, in order to provide the process gas unobstructed access to the tubes.

As indicated in Fig. 4, the gas inlet hood 12 may in addition to the insulation of the tube plate at the gas inlet side contain a filler 54 of ceramic materials, a wire mat mesh or similar material. Furthermore, the gas inlet hood 12 may be made of stainless steel or may have a reaction-inhibiting coating on the inside. Finally, by polishing of the interior surface, the accumulation of catalytic dust blown in from the tubes may be made more difficult.

Fig. 5 shows, in this aspect similar to Figures 1 and 2, a tube plate 60 that is insulated at the heat carrier side.

However, in this case the heat insulation layer consists of a chamber 64 that is closed against the reaction zone 67 of the reactor. The chamber 64 exhibits inlets and outlets 66 and 68 one above the other for a cooling medium as well as a guide plate 70 between the inlet and outlet that forces the cooling medium to flow along the tube plate 60 as well as the separator disk 72 that separates the chamber 64 from the reaction zone 62. The pipes, such as 16, for example, penetrate the separator disk 72 in a sealed manner.

The respective cooling medium can be made of the same or a different medium as the heat carrier in the reaction zone 62. In the first case, it may be diverted at a suitable location after the re-cooling from the heat carrier circulation according to Fig. 1. Also, possible smaller leakages at the tube penetrations through the separator disk 72 are not crucial. Still, with regard to the reaction zone 62, about the same pressure should be maintained in the chamber 64 in order to keep leakage streams at the tube penetrations to a minimum.

The chamber 64 can also be evacuated or filled with an immovable solid, liquid or gaseous heat insulation medium, such as sand, oil or air, for example. In this regard, a liquid or gaseous heat insulation medium can be prevented from circulating through an installed cell structure. In any case, the cooling or heat insulation medium used in the chamber 64 should be one that is incapable of reacting with the heat

carrier used in the reaction chamber 62.

Basically, such chambers can be used at the gas inlet side as well as the heat carrier side of the tube plate on the gas inlet side and may, again, only stretch across part of the tube plate, such as the tube-free zone or the edge zone.

According to Fig. 6, a heat insulation layer 80 on the heat carrier side of a tube plate 82 on the gas inlet side simply consists of a stream-calming zone of the heat carrier that is the result of installations 84 in the shape of honeycombs or concentric ring structures, where the heat carrier will assume a lower temperature than in the actual reaction zone due to the tube plate being cooled by the incoming process gas, regardless whether the reactor operates in parallel or counter flow mode. This applies even more when the contact tubes are not filled with catalyst all the way to the tube plate.

The installations 84 can, but do not necessarily have to be covered by a plate 86 as indicated in Fig. 6 by a dashed line and can also be sealed towards the tube plate 82.

Basically, the invention can be applied to exothermally as well as endothermically operating reactors or multi-step reactors, such as the one shown in Fig. 5 of DE 22 01 528 C regardless whether the gas inlet is on the top or bottom side and the heat carrier passes through the reactor in parallel

flow or counter flow mode.

It generally applies that the tube ends at the tube plate on the gas inlet side can be entirely or in part kept free of catalytic mass or can be filled with an inert material or a mixture of such a catalytic material in order to limit the reaction temperature close to the tube plate.

Patent Claims

1. Tubular reactor (2) for catalytic reactions with a heat carrier that inside a reactor jacket (10) circulates around a contact tube bundle (8), which extends between a tube plate (2; 60; 82) at the reaction gas inlet side and a tube plate (6) at the reaction gas outlet side, as well as gas inlet and gas outlet hoods (12, 14) spanning the face sides of the two tube plates and containing reaction-inhibiting media in the zone of the tube plate on the inlet side of the reaction gas, characterized in that the reaction-inhibiting media consist entirely or in part of a heat insulation layer (46; 50; 64; 80) with openings for the tube cross-sections on at least one of the two sides of the respective tube plate (4; 60; 82).
2. Tubular reactor (2) as set forth in claim 1, characterized in that the heat insulation layer (46; 64; 80) is, at least predominantly, located at the heat carrier side of the tube plate (4; 60; 82).
3. Tubular reactor (2) as set forth in claim 1, characterized in that the heat insulation layer (50) is, at least predominantly, located at the reaction gas inlet side of the tube plate (4).
4. Tubular reactor (2) as set forth in one of the previous

claims, characterized in that the heat insulation layer (46; 50) exhibits a locally varying thickness.

5. Tubular reactor (2) as set forth in one of the previous claims, characterized in that the heat insulation layer (46; 50) exhibits a locally varying composition.

6. Tubular reactor (2) as set forth in one of the previous claims, characterized in that the heat insulation layer (46; 50; 64; 80) is limited to partial zones, such as tube-free areas or the edge area of the tube plate on the gas inlet side (2; 60; 82), for example.

7. Tubular reactor (2) as set forth in one of the previous claims, characterized in that the heat insulation layer (46; 50) consists, at least partially of ceramics, such as a glass frit, for example, or of a respective heat resistant solid material.

8. Tubular reactor (2) as set forth in one of the previous claims, characterized in that the heat insulation layer consists, at least partially, of a chamber (64).

9. Tubular reactor (2) as set forth in claim 8, characterized in that the chamber (64) contains a solid, liquid or gaseous heat insulation material.

10. Tubular reactor (2) as set forth in claim 9, characterized in that a liquid or gaseous heat insulation material in the chamber (64) is prevented from circulating by structures that are installed in the chamber.
11. Tubular reactor (2) as set forth in claim 9, characterized in that a liquid or gaseous cooling medium is circulated throughout the chamber (64).
12. Tubular reactor (2) as set forth in claim 11, characterized in that a partial stream of the heat carrier circulating around the contact tube bundle (8) finds use as a liquid cooling medium.
13. Tubular reactor (2) as set forth in claim 8, characterized in that the chamber (64) is evacuated.
14. Tubular reactor (2) as set forth in one of the previous claims, characterized in that the heat insulation layer (80) consists, at least in part, of a flow-calming zone of the heat carrier due to installations (84) such as honeycomb or concentric ring structures, for example.
15. Tubular reactor (2) as set forth in claim 14, characterized in that the installations (84) are, at least on the side opposite the tube plate (82), covered, preferably sealed.

16. Tubular reactor (2) as set forth in one of the previous claims, characterized in that the gas inlet hood (12) is made of stainless steel.

17. Tubular reactor (2) as set forth in one of the previous claims, characterized in that the interior wall of the gas inlet hood (12) is polished or exhibits a reaction-inhibiting coating.

18. Tubular reactor (2) as set forth in one of the previous claims, characterized in that the gas inlet hood (12) contains a gas-permeable filler (54) of ceramic material, wire mat mesh or similar material.

Abstract

A tubular reactor (2) for catalytic reactions with a heat carrier that inside a reactor jacket (10) circulates around a contact tube bundle (8), which extends between a tube plate (4; 6; 80; 82) at the reaction gas inlet side and one at the reaction gas outlet side with gas inlet and gas outlet hoods (12, 14) spanning the face sides of the two tube plates and containing reaction-inhibiting media in the zone of the tube plate on the gas inlet side, characterize themselves in that the reaction-inhibiting media consist entirely or in part of a heat insulation layer (46; 50; 64; 80) with openings for the tube cross-sections on at least one of the two sides of the respective tube plate (4; 60; 82). In this manner, either the respective tube plate (4; 60; 82) is insulated against the hot heat carrier or the reaction gas entering into the reactor is prevented from having contact with the comparatively hot tube plate in order to prevent harmful secondary reactions at the reactor inlet.

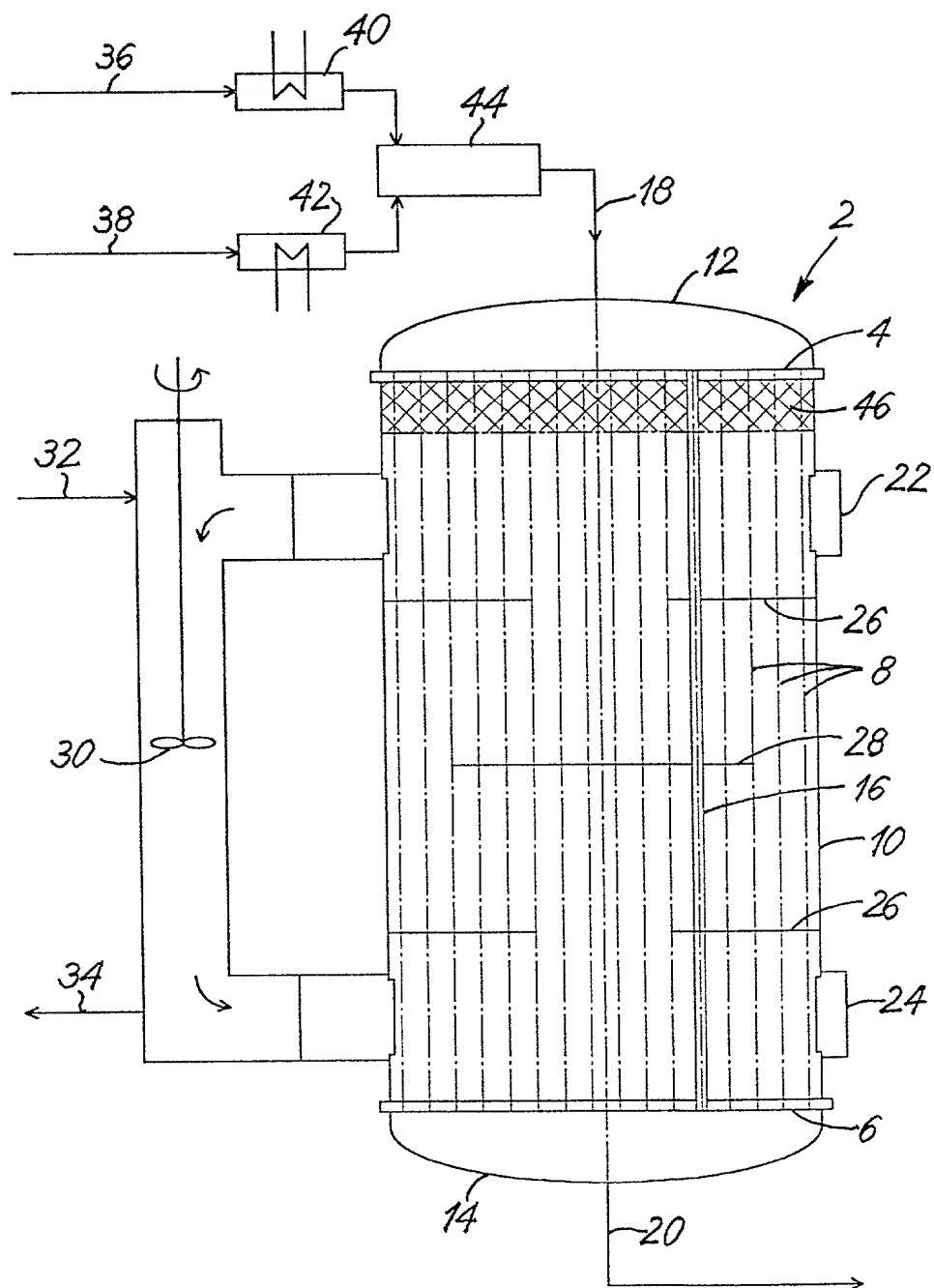


Fig. 1

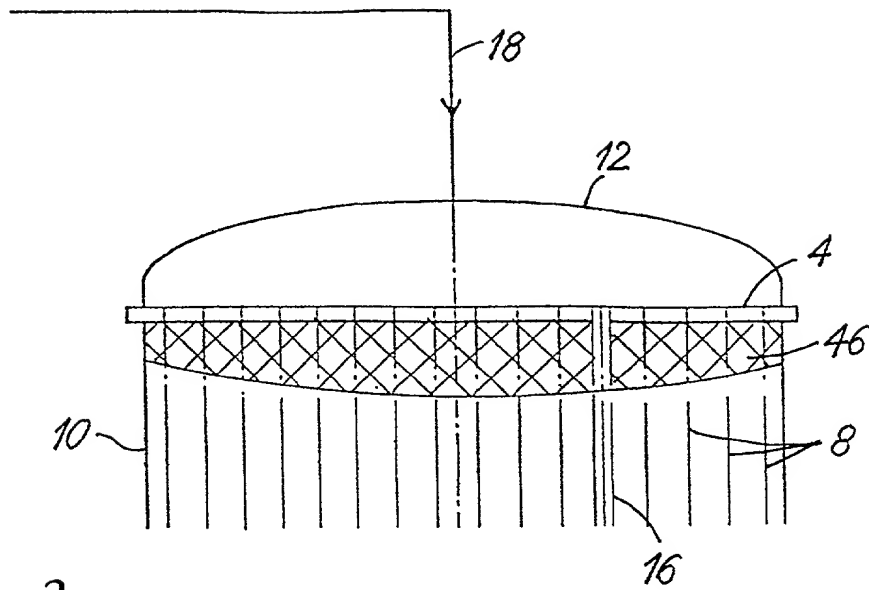


Fig. 2

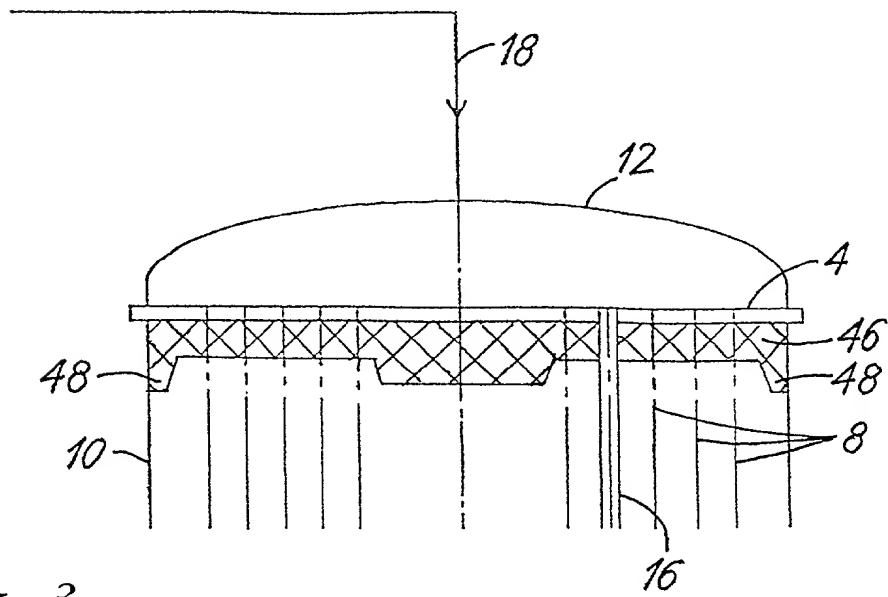
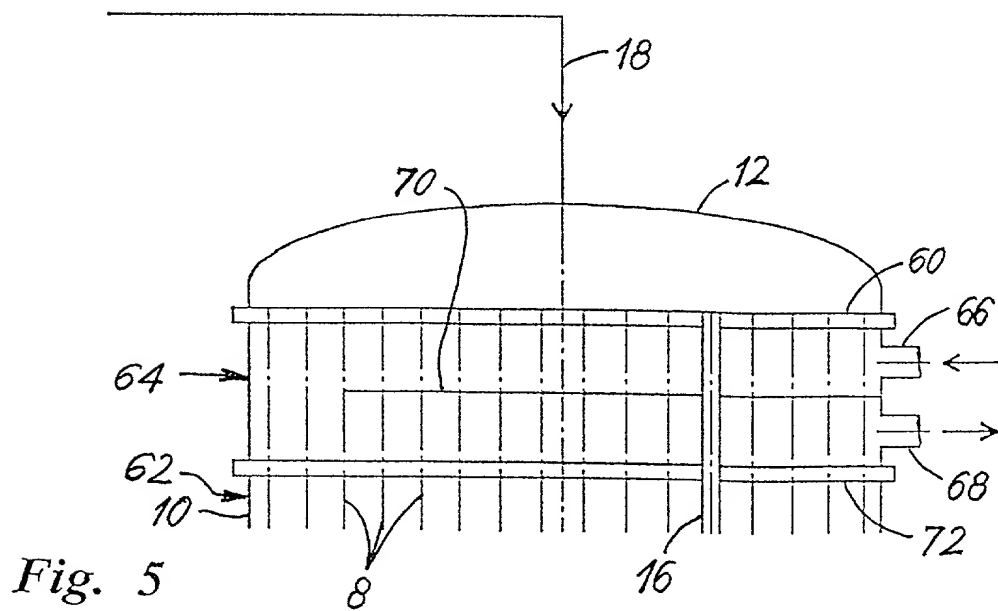
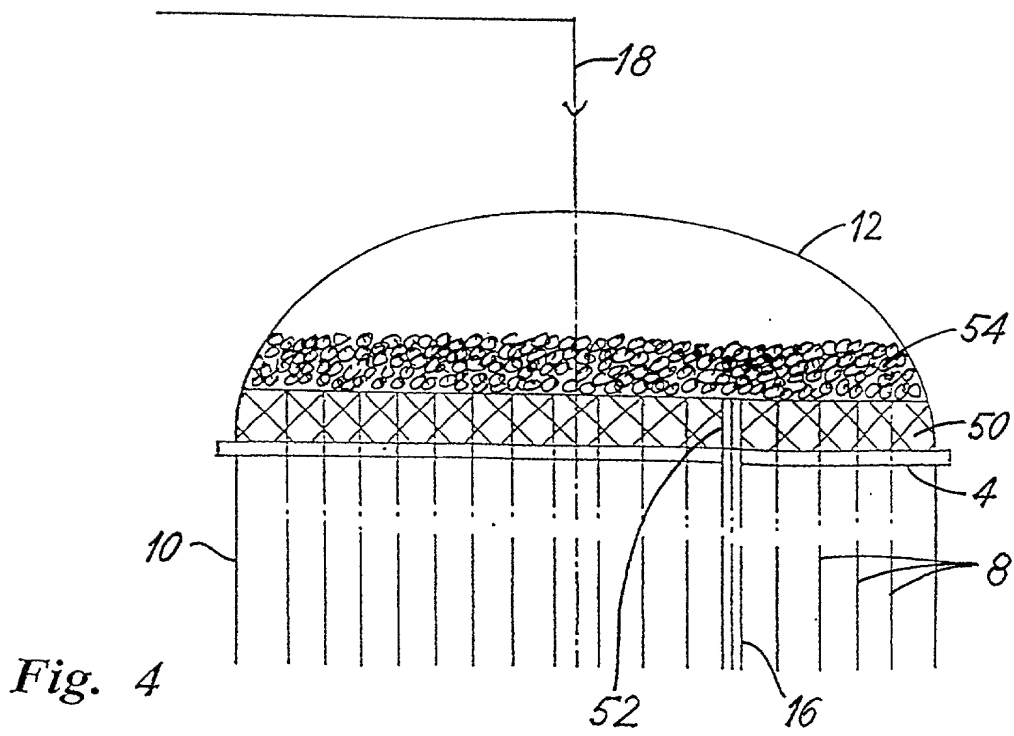
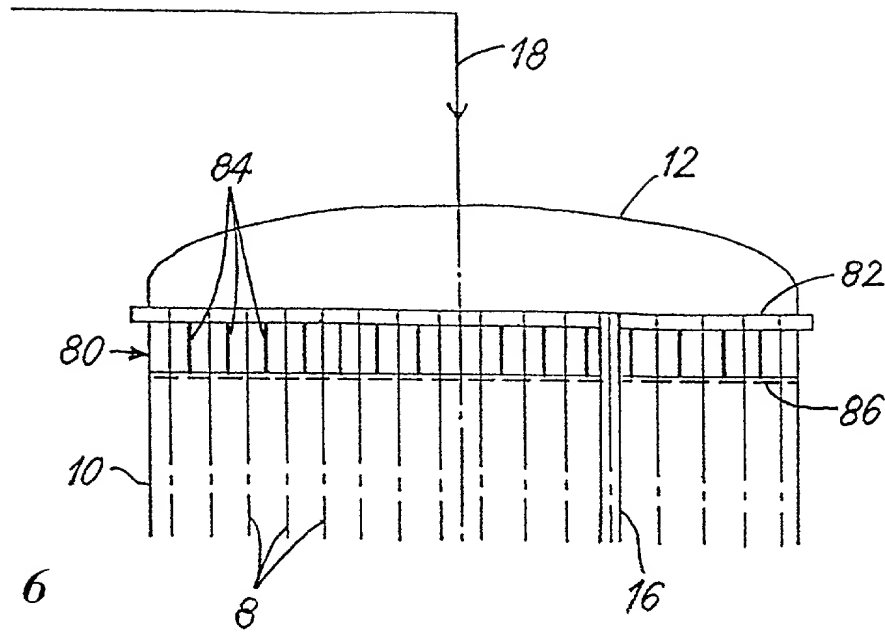


Fig. 3

3/4



4/4

**Fig. 6**

DECLARATION AND POWER OF ATTORNEY
FOR PATENT APPLICATION

Attorney Docket No.
BAUER 214-KFM
D-10826

As the below named inventors, I/We hereby declare that:

My/Our residence, post office address and citizenship is as stated below next to my/our name.

If one name appears below, I am the sole inventor of the subject matter sought to be patented.

If two or more names appear below, we are joint inventors of the subject matter sought to be patented.

I/We believe I/We am/are the original; and first inventor(s) of the subject matter which is claimed and for which a patent is sought on the invention entitled.

TUBULAR REACTOR FOR CATALYTIC REACTIONS

the specification of which

☒ is attached hereto.

☐ was filed on _____ as application Serial No. _____

I/We hereby state that I/We reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I/We acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, Section 1.56(a).

I/We also acknowledge the duty to disclose information which is material to the examination of this application in accordance with Title 37, Code of Federal Regulations, Section 1.63(d), which occurred between the filing date of the prior application and the filing date of the continuation-in-part application, if this is a continuation-in-part application.

I/We hereby claim foreign priority benefits under Title 35, United States Code, Section 119 of any foreign application(s) for the patent or inventor's certificate listed below and have also identified below any foreign application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

Prior Foreign Application: GERMAN Application No. 198 06 810.7
filed February 18, 1998

Priority Claimed: x Yes No

Prior Foreign Application: PCT Application No. PCT/EP99/01036
filed February 17, 1999

Priority Claimed: x Yes No

I/We hereby claim the benefit under Title 35, United States Code, Section 120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code, Section 112, I acknowledge the duty to disclose material information as defined in Title 37, Code of Federal Regulations, Section 1.56(a) which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

| <u>Application Serial No.</u> | <u>Filing Date</u> | <u>Status</u> (patented, pending, abandoned) |
|-------------------------------|--------------------|---|
|-------------------------------|--------------------|---|

I/We hereby declare that all statements made herein of my/our own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

I/We hereby appoint the following attorneys and/or agents to represent me with respect to the above identified U.S. Patent Application, and to prosecute any continuations, continuations-in-part, reissue applications and/or reexaminations with respect to these applications and to transact all business in the Patent and Trademark Office connected therewith, and hereby expressly revoke all prior powers, whatever they may be, heretofore had herein:

3- Karl F. Milde, Jr., Reg. No. 24,822; Steven M. Hoffberg, Reg. No. 33,511 and Kenneth E. Macklin, Reg. No. 20,875, all of 10 Bank Street, Suite 460, White Plains, New York 10606, my/our attorneys with full power of substitution and revocation.

Please address all telephone calls to Karl F. Milde, Jr., Esq. at telephone No. (914) 949-3100.

Please address all correspondence to:

Karl F. Milde, Jr., Esq.

MILDE, HOFFBERG & MACKLIN, L.L.P.

10 Bank Street - Suite 460

White Plains, New York 10606

1-80
Friedrich Guetlhuber
NAME OF INVENTOR

Friedrich Guetlhuber
INVENTOR'S SIGNATURE

July 10, 2000
DATE

Gartenstrasse 4, D-94526 Metten, GERMANY
RESIDENCE

DEX

GERMAN
CITIZENSHIP

SAME AS ABOVE
POST OFFICE ADDRESS

BAUER 214-KFM:jg
D-10826

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : JOHANN ORTNER and JOHANN SEEMANN
Serial No.: 09/622,301
Filed : August 16, 2000
For : TUBULAR REACTOR FOR CATALYTIC REACTIONS

October 25, 2000

Hon. Commissioner of Patents
& Trademarks
Washington, DC 20231

Attn.: Ms. Paulette Kidwell, Paralegal - 703 305-3230

Sir:

TRANSMITTAL OF ENGLISH TRANSLATION

Attached herewith is a copy of applicants' translation
of the original International Application No.

PCT/EP99/01036.

Please charge the surcharge in the amount of \$130 (for
late filing of translation) to our Deposit Account No. 50-
0427.

Respectfully submitted,

By

Karl F. Milde, Jr.
Reg. No. 24,822

MILDE, HOFFBERG & MACKLIN, LLP
10 Bank Street - Suite 460
White Plains, NY 10606

(914) 949-3100

I hereby certify that this correspondence,
16 pages, is being facsimile transmitted
to The Commissioner of Patents and Trademarks
Washington, D.C. 20231 on the date shown below.

By

10-25-00
J. Milde, Jr.